PHYSICAL REVIEW B VOLUME 10, NUMBER 1 1 JULY 1974

Determination of exchange integrals J_1 and J_2 and magnetic surface-anisotropy energy in EuS from standing-spin-wave resonance*

Peter K. Schwob[†], M. Tachiki[‡§], and Glen E. Everett
Physics Department, University of California, Riverside, California 92502

(Received 2 October 1972)

The standing-spin-wave absorption spectrum in thin EuS films has been measured in the temperature interval 1.3–4.2 K. The dependence of the magnetization on temperature has been determined from the ferromagnetic-resonance field and compared with spin-wave theory. The results are well described by the exchange-parameter combination $(J_1 + J_2)/k_B = (0.096 \pm 0.003)$ K. The temperature dependence of the exchange stiffness parameter $D(T)$ has been calculated including the contribution of a nonzero internal field. The experimental results have been analyzed taking the surface boundary conditions into account and correcting for the temperature and field dependence of H_i. The results show that the apparent temperature dependence of $D(T)$ arises almost entirely from the surface-anisotropy energy. The small observed variation $\Delta D(T)/D_0$ has been analyzed yielding $J_2/J_1 = -0.57 \pm 0.05$. The results, expressed as $J_1/k_B = (0.214 \pm 0.026)$ K, $J_2/k_B = -(0.122 \pm 0.025)$ K are in essential agreement with the inelastic-neutron-scattering determination of Passell *et al.* but in marked disagreement with Swendsen's Green's-function theory and its application to the calculation of the ferromagnetic and paramagnetic Curie temperature. A comparison with other experimental determinations of J_1 and J_2 is made.

I. INTRODUCTION

The existence of an "ideal Heisenberg ferromagnet" was recognized with the discovery of ferromagnetism in the semiconducting divalent europium compounds EuO[1] and EuS.[2-6] The half-filled $4f$ shell of the Eu^{2+} ion is described by a localized effective-spin ground state $^8S_{7/2}$ for which the exchange energies are very small compared to the energy of a possible excited state having a different effective spin.[3(b),6] The S-state character is expected to lead to very small magnetocrystalline anisotropy energies which has been verified experimentally.[7,8] Evidence for a weak pseudo-dipolar interaction in EuO is obtained from the high-temperature paramagnetic-resonance linewidth[9] and the temperature dependence of the second-order magnetocrystalline anisotropy constant K_2.[10] The temperature dependence of the first- and second-order anisotropy constants in EuS[8(b)] can be described with just single-ion and dipolar contributions[10] thus more nearly representing the "ideal Heisenberg ferromagnet." The NaCl structure of the europium chalcogenides together with the evidence that only nearest-neighbor J_1 and next-nearest-neighbor J_2 exchange interactions are important[2(b)] makes the calculation of the spin-wave excitation energies straightforward.

Spin-wave theory has been used by Callaway and McCollum[11] to interpret their low-temperature specific-heat measurements in EuS, employing the "exact dispersion relations" and integrating numerically over the entire Brillouin zone (BZ) but neglecting dipolar and magnon-magnon interactions. The theoretically predicted contributions to the specific heat proportional to $T^{3/2}$ and $T^{5/2}$ were observed and from their coefficients, the values of J_1 and J_2 were determined. Charap and Boyd[12] using NMR, measured the zero-field magnetization as a function of temperature and analyzed their results together with the specific-heat data of Callaway and McCollum,[11] using the dispersion relations calculated including the dipolar interaction as first determined by Holstein and Primakoff.[13] Charap and Boyd[12] introduced parametrically a magnetic field H_a to account for the presence of domains at zero applied field. By adjusting the values of J_1, J_2, and H_a to give the best agreement with the experimental results, they found a "range" of values for these parameters which fit the data equally well selecting the set

$$J_1/k_B = (0.20 \pm 0.01)\ \text{K};$$

$$J_2/k_B = -(0.08 \pm 0.02)\ \text{K};$$

$$H_a = 4 \pm 2\ \text{kOe}.$$

The value of $H_a \approx 4$ kOe, introduced to represent the anisotropy fields in the domain walls, is to be compared with the measured values $K_1/M \sim -30$ Oe and $K_2/M \simeq 12$ Oe in the temperature interval of their analysis.[8(b)] Passenheim, McCollum, and Callaway[14] have measured the specific heat in the presence of an externally applied field. They used the "exact" dispersion relation following Charap and Boyd[12] but with the actual values for H_i, rather than using this quantity as an adjustable parameter. The values of J_1/k_B and J_2/k_B that gave the best fit to the data were 0.20 and −0.06 K, respectively. With this choice, the fit of the experimental data in zero field is reasonably good but becomes progressively worse at the higher fields. They state that at their highest internal field, $H_i = 10.8$ kOe, different choices of J_1 and J_2 would give agreement over different limited temperature ranges, but with no single set eliminating the systematic deviation at either the low- and/or high-temperature limits of the measured temperature interval.

A more critical test of spin-wave theory is afforded by the direct determination of the dispersion relations made possible by inelastic-neutron-scattering and standing-spin-wave-resonance (SSWR) experiments. The former, when performed on single crystals, is capable of yielding the dispersion relation over the entire BZ but becomes insensitive for low k values near the zone center. The complimentary SSWR experiments are most sensitive for small values of k. We have performed SSWR measurements on thin films of EuS and reported the value of the combination $J_1 + J_2$ determined at one temperature.[15] Inelastic-neutron-scattering measurements on polycrystalline EuS have also been performed.[16] We shall defer a comparison of the results obtained by the various types of experiment to the discussion section noting here only that a small disagreement continues to exist between SSWR and neutron scattering results.

We have extended our previous work at 1.1 K to the interval 1.1–4.2 K measuring the SSWR absorption as a function of field at constant frequency. An analysis of our data, to yield reliable results for the exchange constants, requires that the variation of the magnetization with field be corrected for and that the spin-wave boundary condition be properly included. The procedure employed was as follows:

(1) The ferromagnetic-resonance or uniform-precession field is given by

$$\omega/\gamma = H - 4\pi M.$$

The temperature dependence of the resonant field was determined giving the corresponding variation of $M(H,T)$.

(2) The variation of $M(H,T)$ was analyzed using spin-wave theory yielding a determination of the linear combination $J_1 + J_2$. The dispersion relations used include the Zeeman and dipolar contributions. In Sec. II, the expression for $M(H,T)$ is given as an integral over the BZ and simplified for purposes of numerical analysis by replacing the BZ with an "equivalent spherical volume."

(3) Spin-wave theory was used with the value of $J_1 + J_2$ determined to calculate $M(H,T)$ at each peak position in the observed absorption yielding the absorption as a function of internal field.

(4) The SSWR absorption was analyzed by including a surface-anisotropy energy to describe the incomplete spin pinning at the surfaces. The wave-vector dependence of the boundary conditions was used explicitly. The transcendental equations (described in the Appendix) were solved numerically to give a best fit to the data. This analysis gives an independent determination of $J_1 + J_2$. The "exchange stiffness" and the surface-anisotropy energy were determined in this way at each temperature.

(5) The temperature dependence of the exchange stiffness, $D(T)$, has been analyzed using spin-wave theory. In Sec. II we obtain expressions for $D(T)$ by taking into account the contribution of magnon-magnon interactions to the dispersion relations and the orientation dependence of this term in single-crystal samples. The Zeeman and dipolar contributions to the dispersion relations were included and the resulting integrals simplified by replacing the BZ with "an equivalent spherical volume."

In what follows we give, Sec. II, an outline of the spin-wave calculations and the expressions used in our analysis. Details regarding preparation of our samples is given in Sec. III followed by our results and their analysis in Sec. IV. We compare our results with those from other experiments in Sec. V concluding that the various experimental determinations of J_1 and J_2 are in good agreement. Comparison with a Green's-function theory which calculates the paramagnetic and ferromagnetic Curie temperatures indicates a significant disagreement between theory and experiment.

II. THEORY

We shall represent the Hamiltonian of our system as

$$\hat{H} = \hat{H}_{\text{ex}} + \hat{H}_d + \hat{H}_Z, \tag{1}$$

where

$$\hat{H}_{\text{ex}} = -\frac{1}{2} \sum_{\substack{l,m \\ (l \neq m)}}^{N} 2J(R_{lm}) \vec{S}_l \cdot \vec{S}_m \tag{2a}$$

and is of the Heisenberg form. The exchange integrals, $J(R_{lm}) \equiv J_{lm}$ represent the coupling between atoms at lattice sites l and m separated by the distance R_{lm}. The dipolar $\hat{H}_d$ and Zeeman $\hat{H}_Z$ contributions can be represented by

$$\hat{H}_d = \frac{1}{2} \sum_{\substack{l,m \\ (l \neq m)}}^{N} \frac{(g\mu_B)^2}{R_{lm}^3} \times \left[\vec{S}_l \cdot \vec{S}_m - \frac{3}{R_{lm}^2} (\vec{S}_l \cdot \vec{R}_{lm})(\vec{S}_m \cdot \vec{R}_{lm}) \right], \tag{2b}$$

$$\hat{H}_Z = -g\mu_B \vec{H} \cdot \sum_{l}^{N} \vec{S}_l \,. \tag{2c}$$

The inclusion of terms representing single-ion anisotropy would lead to an additional contribution to the spin-wave energy gap which for our experimental conditions is negligible compared to that introduced by the external field and hence can be neglected. The g factor is a scalar having the value $g \cong 2$ in the europium chalcogenides.

The excitation energies $\hbar\omega_k$ for the Hamiltonian, Eq. (1), were calculated first for a needle-shaped sample by Holstein and Primakoff[13] and extended to the general ellipsoid by Anderson and Suhl.[17] For cubic crystals the result is[18]

$$\hbar\omega_k = \{ g\mu_B H + 2S(J_0 - J_k) + [4\pi - N_z - 4\pi(k_z/k)^2] g\mu_B M \}^{1/2} \times \{ g\mu_B(H - N_z M) + 2S(J_0 - J_k) \}^{1/2}, \tag{3}$$

where

$$J_k = \sum_{\substack{m \\ (m \neq l)}} J_{lm} e^{i \vec{k} \cdot \vec{R}_{lm}}, \tag{4}$$

and N_z is the demagnetizing factor in the direction of the applied field. For the field normal to the surface of a thin film, i.e., an infinite plate, $N_z = 4\pi$. Expressed in terms of the internal field, $H_i = H_0 - 4\pi M$, Eq. (3) becomes

$$\hbar\omega_k = \{ g\mu_B H_i + 2S(J_0 - J_k) + 4\pi g\mu_B M \sin^2\theta_k \}^{1/2} \times \{ g\mu_B H_i + 2S(J_0 - J_k) \}^{1/2}, \tag{5}$$

where θ_k is the angle between the wave vector $\vec{k}$ of the excitation and the applied field direction.

We can estimate the relative importance of these contributions to the excitation energy by first observing that the exchange related energy, $2S(J_0 - J_k)$, goes from zero at the center to a maximum at the corners of the Brillouin zone, with the maximum value being given approximately by $k_B T_C$, where T_C is the Curie temperature. The Zeeman- ($H = 8.6$ kG, typically) and dipolar- ($4\pi M = 15.2$ kG in EuS at $T = 0$ K) related energies expressed in temperature units are ~1 and 2 K, respectively, and are not negligible compared to the maximum exchange contribution, $T_C = 16.5$ K. Moreover, we shall be interested in temperatures $T \lesssim 0.25 T_C$ for which thermally excited spin waves with only small k will be important. Hence the Zeeman, dipolar, and exchange terms will be of equal importance in calculating thermodynamic properties. In the calculation of magnetization and exchange stiffness constants which follow, we shall use the excitation energies given by Eq. (5) but expanding $2S(J_0 - J_k)$ to first order in k^2. Further, we shall limit our summations over k to the interval 0 to k_c, where k_c is the radius of a sphere having the same volume as the BZ. We feel that these approximations, while seeming to be more restrictive than the exact numerical integration of Eq. (5) over the entire BZ and not expanding $2S(J_0 - J_k)$[12,14] are likely to be just as reliable as the "exact" integrations, since it is known that replacement of the dipolar sums by integrals in obtaining Eq. (5) is not appropriate for values of k near the BZ boundaries.[19]

A. Temperature and field dependence of the intrinsic domain magnetization

The temperature and field dependence of the intrinsic domain magnetization for temperatures $T/T_C \ll 1$ has been calculated by Holstein and Primakoff.[13] Briefly, the free energy, $F(H, T)$, is calculated by taking the expectation value of the diagonalized Hamiltonian and the magnetization calculated from the relation $M = -(\partial F/\partial H)_T$. The result is

$$M(H,T) = g\mu_B NS - g\mu_B \sum_k \frac{A_k}{\hbar\omega_k} \langle \hat{n}_k \rangle - \frac{g\mu_B}{2} \sum_k \left[\frac{A_k}{\hbar\omega_k} - 1 \right] + O(\langle \hat{n}_k \rangle^2), \tag{6}$$

where

$$A_k = 2S(J_0 - J_k) + 2\pi g\mu_B M \sin^2\theta_k + g\mu_B H_i$$

and

$$\langle \hat{n}_k \rangle = [e^{\hbar\omega_k / k_B T} - 1]^{-1}.$$

The terms $O(\langle \hat{n}_k \rangle^2)$ will be small at low temperatures but of comparable magnitude to those terms neglected by Holstein and Primakoff[13] in diagonalizing the Hamiltonian. For consistency, these terms will be neglected. The summation in Eq. (6) is over the first BZ and N is the number of magnetic ions per unit volume. The first three terms in Eq. (6) can be written

$$M(H,T) = M_0 - \mathfrak{M}(H,T) - \mathfrak{M}(H). \tag{7}$$

In this expression M_0 is the magnetization that would result for complete spin parallelism. The small reduction in ferromagnets of this pseudoclassical result at zero temperature due to zero-point effects is given by $\mathfrak{M}(H)$. The decrease in the magnetization at finite temperatures due to spin-wave excitations is given by $\mathfrak{M}(H,T)$. Since we shall use this expression to (i) analyze our results for the variation of M with temperature at constant internal field and (ii) obtain the small correction to the internal field necessary in analyzing the SSWR results, it will be sufficient to regard $M_0 - \mathfrak{M}(H)$ as being a constant and evaluate just $\mathfrak{M}(H,T)$.[20]

The evaluation of $\mathfrak{M}(T,H)$ is performed by replacing the sum by an integral. We expand the quantity $2S(J_0 - J_k)$ in k^2 obtaining

$$2S(J_0 - J_k) = 2S(J_1 + J_2)a^2k^2 + O(k^4), \tag{8}$$

where J_1 and J_2 are the first- and second-nearest-neighbor exchange integrals. The contribution of more distant neighbors could be included [see Eq. (4)], but for EuS there is presently no evidence indicating that such a possibility should be considered. The effect of including the term in k^4 would be to multiply the first term in Eq. (8) by $1-(\frac{1}{6}ak)^2$ and since we shall only be concerned with temperatures much smaller than the Curie temperature, can be neglected. Our final simplifying approximation is the replacement of the BZ by a spherical volume of radius k_c chosen such that the two volumes are equal. This value of k_c is given by $k_c^3 = 6\pi^2 N$, where N is the number of magnetic atoms per unit volume. For the fcc lattice, k_c is given by

$$k_c = \frac{2\pi}{a}(3/\pi)^{1/3}. \tag{9}$$

We note that these approximations are the same as those used in the Debye model for lattice vibrations. By writing

$$x = \cos\theta_k, \quad p = ak, \quad A = 2S(J_1 + J_2),$$

we obtain the following expression for $\mathfrak{M}(H,T)$:

$$\mathfrak{M}(H,T) = \frac{g\mu_B N}{8\pi^2}\int_0^{p_c} p^2\,dp \times \int_0^1 \frac{Ap^2 + g\mu_B H_i + 2\pi g\mu_B M_0(1-x^2)}{\hbar\omega(e^{\hbar\omega/k_B T} - 1)}\,dx \tag{10a}$$

with

$$\hbar\omega = \{(Ap^2 + g\mu_B H_i) \times [Ap^2 + g\mu_B H_i + 4\pi g\mu_B M_0(1-x^2)]\}^{1/2}. \tag{10b}$$

This expression is approximate but results in a substantial simplification in the subsequent numerical integrations required for comparison with the experimental results. We emphasize again that the expression for $2S(J_0 - J_k)$ used in "exact numerical integration over the BZ"[12,14] becomes an approximation for values of k near the BZ boundaries. We feel that unless more accurate expressions for $\hbar\omega_k$ near the BZ boundaries were used in calculating $\mathfrak{M}(H,T)$, the increased complication of "exact integration" is not required for a comparison with experimental data.

B. Temperature and field dependence of the exchange stiffness constant

It has been shown that the temperature dependence of the exchange stiffness constant originates from the interaction between spin waves, and is proportional to $T^{5/2}$ in the case of vanishing dipolar and Zeeman interactions.[18] For EuS the temperature and fields we consider are such that the dipolar, exchange, and Zeeman interactions are of comparable importance in determining the spin-wave spectrum and the resulting temperature dependence of the exchange stiffness constant differs from the well-known $T^{5/2}$ result. In calculating the noninteracting spin-wave spectrum, the dipolar interaction contributes a wave-vector-independent term proportional to $4\pi M$. The magnon-magnon interactions will contain contributions from both dipolar and exchange terms neglected in the Holstein-Primakoff expansion.[13] A comparison of the relative contribution to the coefficient of a general term in four spin-wave operators indicates that the dipolar part is much weaker than that of exchange. We will assume that this relative contribution to the individual coefficients will be the same as the contributions to the renormalized-spin-wave energies. We will, therefore, neglect dipolar terms and consider only the magnon-magnon-interaction terms arising from exchange.

We have calculated the temperature dependence of the exchange stiffness constant by the following procedure.[21] We expand the exchange interaction using the Holstein-Primakoff method in a power series of boson operators. The second-order terms have already been taken into account in the spin-wave energy, Eq. (3). The fourth-order term is given by

$$-\frac{1}{2N}\sum_{k_1}\sum_{k_2}\sum_{k_3}\sum_{k_4}(2J_{k_1-k_3} - J_{k_1} - J_{k_3})a^\dagger_{k_1}a^\dagger_{k_2}a_{k_3}a_{k_4} \times \delta(k_1 + k_2 - k_3 - k_4), \tag{11}$$

where J_k has been defined by Eq. (4), and $a^\dagger_k$ and a_k are boson creation and annihilation operators, respectively. Terms higher than fourth order will be neglected. We now apply the same transformations, i.e., the second and third Holstein-Primakoff transformations, that were used in diagonalizing the Hamiltonian when only second-order terms in the spin-wave operators were considered. The off-diagonal terms give rise to magnon-magnon scattering and contribute to the life-time broadening of the spin-wave spectrum. For the temperature shift of the spin-wave energies we need only retain those terms from Eq. (11) that are diagonal. With this approximation, the total energy for the spin-wave excitation energy becomes

$$E=\sum_{k_1}\hbar\omega_{k_1}\hat{n}_{k_1}-\sum_{k_1k_2}I_{k_1k_2}\hat{n}_{k_1}\hat{n}_{k_2}\,, \tag{12}$$

where $I_{k_1k_2}$ represents the interaction between the spin waves of wave vector k_1 and k_2, and $\hat{n}_{k_1}$ and $\hat{n}_{k_2}$ are their occupation numbers. The energy required to create a spin wave of wave vector k_1, $\hbar\Omega_{k_1}$, is given by

$$\hbar\Omega_{k_1}=\hbar\omega_{k_1}-2\sum_{k_2}I_{k_1k_2}\langle\hat{n}_{k_2}\rangle\,, \tag{13}$$

where $\hat{n}_{k_2}$ has been replaced by the thermal expectation value. We shall use this result in analyzing our SSWR results and need only consider those values of k_1 in Eq. (13) which are parallel to the applied field, i.e., the z axis. With this restriction $I_{k_1k_2}$ can be written

$$I_{k_1k_2}=-\frac{1}{N}(J_0+J_{k_1-k_2}-J_{k_1}-J_{k_2})\frac{A_{k_2}}{\hbar\omega_{k_2}}\,. \tag{14}$$

We expand the term $(J_0+J_{k_1-k_2}-J_{k_1}-J_{k_2})$ in terms of k_1 and k_2 and observe that the mixing from the term $J_{k_1-k_2}$ will be sensitive to the crystallographic direction along which H is applied. The actual EuS films produced appear to crystallize with definite crystallographic planes being preferred, hence we will consider two specific cases in expanding this term. We obtain the results

$\vec{\mathrm{H}}\parallel\langle 100\rangle$:

$$(\tfrac{1}{16}J_1(k_{2z}^2+k_2^2)+\tfrac{1}{2}J_2k_{2z}^2)a^4k_{1z}^2\;;$$

$\vec{\mathrm{H}}\parallel\langle 111\rangle$:

$$[\tfrac{1}{12}(J_1+2J_2)k_2^2]a^4k_{1z}^2\;.$$

At zero temperature $\hbar\Omega_k=\hbar\omega_k$ since $\langle\hat{n}_{k_2}\rangle$ is zero. We define the temperature-dependent part of the exchange stiffness constant, $\Delta D(T)$, by the relation

$$\Delta D(T)k_1^2=\hbar\Omega_{k_1}-\hbar\omega_{k_1}\,. \tag{15}$$

We use this definition obtaining for the two cases

$\vec{\mathrm{H}}\parallel\langle 100\rangle$:

$$\Delta D(T)=\frac{-a^4}{N}\sum_k\{\tfrac{1}{8}J_1(k^2+k_z^2)+J_2k_z^2\}\frac{A_k}{\hbar\omega_k}\langle\hat{n}_k\rangle\;; \tag{16a}$$

$\vec{\mathrm{H}}\parallel\langle 111\rangle$:

$$\Delta D(T)=-\frac{a^4}{N}\sum_k(\tfrac{1}{6}J_1+\tfrac{1}{3}J_2)k^2\frac{A_k}{\hbar\omega_k}\langle\hat{n}_k\rangle\;. \tag{16b}$$

We introduce the same approximations for A_k, $\hbar\omega_k$, and the volume of integration used in the previous subsection in calculating $\mathfrak{M}(H,T)$, obtaining the following expressions for $\Delta D(T)$ which are evaluated numerically:

$\vec{\mathrm{H}}\parallel\langle 100\rangle$:

$$\frac{\Delta D(T)}{a^2}=-\frac{1}{8\pi^2}\int_0^{p_c}p^4dp\int_0^1\frac{[(\frac{1}{8}J_1)(1+x^2)+J_2x^2][Ap^2+g\mu_BH_i+2\pi g\mu_BM_0(1-x^2)]}{\hbar\omega\{e^{\hbar\omega/k_BT}-1\}}dx\,, \tag{17a}$$

$\vec{\mathrm{H}}\parallel\langle 111\rangle$:

$$\frac{\Delta D(T)}{a^2}=-\frac{J_1+2J_2}{48\pi^2}\int_0^{p_c}p^4dp\int_0^1\frac{[Ap^2+g\mu_BH_i+2\pi g\mu_BM_0(1-x^2)]}{\hbar\omega\{e^{\hbar\omega/k_BT}-1\}}dx\,. \tag{17b}$$

These expressions, in the limit $H_i, M_0=0$, reduce to the expression obtained by Marshall[22] and $\Delta D(T)\sim T^{5/2}$. (It is clear that the $T^{5/2}$ dependence is a consequence of being able to neglect the Zeeman and dipolar contributions to the spin-wave spectrum.) In EuS the three terms are of comparable importance and, in particular, the gap in the excitation spectrum due to the internal field, Eq. (5), precludes a simple power-law dependence on temperature in the general case.

An inspection of Eqs. (17) indicates that $\Delta D(T)$ depends on the combination $A=2S(J_1+J_2)$ in the same way for both cases considered. The variation with temperature for a $\langle 111\rangle$ film depends only on A and is insensitive to the relative values of J_1 and J_2. The coefficient, J_1+2J_2, however, is sensitive to the relative values and, in particular, is zero when $J_2=-(\frac{1}{2}J_1)$. For a $\langle 100\rangle$ film, the relative values of J_1 and J_2 determine the magnitude and temperature dependence of $\Delta D(T)$. We have evaluated Eqs. (17) numerically using $(J_1+J_2)/k_B=0.094$ K and $H_i=8.4$ kG for various values of J_2/J_1, the results are shown in Figs. 1 and 2. The sensitivity of the temperature dependence to the ratio J_2/J_1 in $\langle 100\rangle$ films is illustrated in Fig. 1. For values of J_2/J_1 of about -0.47 these results indicate that $\Delta D(T)$ is initially positive increasing to a weak maximum at

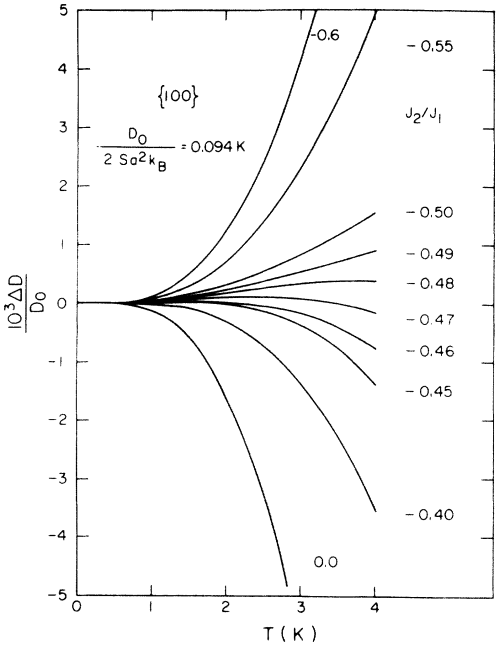

FIG. 1. Calculated variation of the exchange stiffness constant expressed as the relative change $\Delta D(T)/D_0$ versus temperature with an internal field of 8.6 kG directed along a $\langle 100 \rangle$ direction for various values of the ratio J_2/J_1, Eq. (17a).

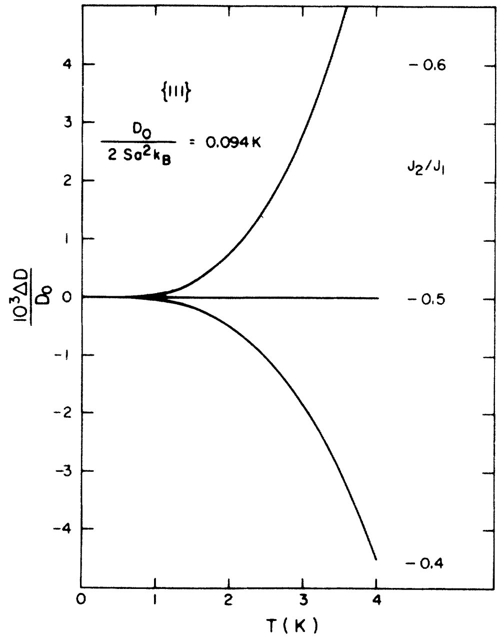

FIG. 2. Same as Fig. 1 but with $\vec{H}_i$ along a $\langle 111 \rangle$ direction Eq. (17b).

~2 K. At higher temperatures, $\Delta D(T)$ decreases and becomes negative. This behavior is the consequence of nearest- and next-nearest-neighbor interactions not being equivalent for spin waves propagating in a $\langle 100 \rangle$ direction. An observation of this effect in a suitable material would be most interesting but is too small for observation in our experiment. We anticipate our experimental results and note that for a $\langle 111 \rangle$ film, J_1 and J_2 contribute in the same way leading to a perfect cancellation at all temperatures when $J_2/J_1 = -0.5$.

III. EXPERIMENTAL DETAILS

Thin films were prepared by vacuum evaporation of EuS onto heated fuzed-quartz substrates. The evaporant was prepared by the H_2S conversion of Eu_2O_3 to EuS followed by a factional distillation in a vacuum furnace to increase the purity. The vacuum during deposition was characteristically $(3-5)\times 10^{-6}$ Torr. Our maximum available substrate temperature of 600 °C yielded high-quality films where the width of the field-normal ferromagnetic (FMR) and observability of SSW modes was used as a criterion. Film thickness was determined to an estimated accuracy of better than 10% by optical interferometry, weight increase determination, and/or an oscillating quartz-crystal thickness monitor. Representative film thicknesses for the SSWR were 250 and 600 A.

An x-ray investigation of film texture in our thinnest film lacking sufficient sensitivity, thicker films were prepared using identical procedures. The results for a 1400-Å film indicated preferential growth in {111} and {311} directions. Indirect evidence for preferential growth in the thinner films is provided by the FMR linewidth. The shift in resonant field due to magnetocrystalline anisotropy was averaged over all crystallographic orientations to estimate the line width in a polycrystalline film. Using the known values of the bulk anisotropy constants at 1.3 K,[23] the polycrystalline average predicts a linewidth of 100 G and is to be compared with the measured value of 40 G at the same temperature. This value in turn is higher than that of bulk single crystals. We conclude that the films used for the SSWR have a crystalline character with [111] and [311] planes being dominant. The field (at constant frequency) for excitation of the nth SSW mode measured with respect to the FMR is inversely proportional to the film thickness squared. By assuming that the "width" of a particular mode is due just to variations in thickness, we estimate a nonuniformity of less than plus or minus four lattice parameters, $\sim\pm 25$ Å. The magnetization determined at our lowest temperature corresponds to a magnetic moment of $6.93\mu_B$/ion compared to the theoretical value of $7.00\mu_B$/ion. We estimate the departure from stochiometry to be no greater than 1%. Inasmuch as the fuzed-quartz substrate contracts less than EuS, this represents an upper limit indicating that the possible formation of nonmagnetic Eu_2O_3 or decomposition of the EuS molecule during evaporation have a negligible effect on the deposited films.

The microwave spectrometer was of standard design employing a resonant cavity, ferrite circulator, and 60-MHz superhetrodyne detection and was operated at 23.6 GHz ± 0.07% as measured with a commercial cavity wavemeter. The signal Klystron was frequency locked to the resonant cavity. The cavity coupling could be adjusted at each operating temperature for optimum sensitivity. The samples were secured in a rectangular cavity using a small quantity of vacuum grease as an adhesive on the substrate side not bearing the Eus film. The sample holder was constructed and the magnetic field subsequently aligned to most nearly achieve the "field-normal" configuration. Thermal contact between the cavity and bath was made using an exchange-gas pressure of 0.5 Torr in the can employed to exclude liquid helium from the cavity interior. The bath temperature was manostatically controlled, but the sample temperature measured in zero field with a Ge resistance thermometer attached to the cavity. The uncertainties in field and temperature measurements are less than ±3 G and 0.05 K, respectively.

The choice of film thicknesses was a trade off between obtaining high resolution for the low-order SSW peaks (thinner) and increasing the total number that could be observed (thicker). For thicknesses greater than 800 Å, the observed absorption spectrum was very different owing to the excitation of coupled magnon-phonon modes. These coupled modes have not been investigated in detail, but an estimate of the dispersion relations using the elastic constants measured at 77 K[24] is consistent with this interpretation. Films thin enough to suppress these modes were used and the need to consider them in our analysis eliminated.

IV. EXPERIMENTAL RESULTS AND ANALYSIS

A. Effective exchange stiffness D at 1.2 K

Our earlier results at 1.1 K[15] have been extended to determine the temperature dependence of the exchange stiffness constant. For complete spin pinning at the film surfaces, the difference between the field for the uniform precession, H_0, and the nth standing spin wave, H_n, measured at constant frequency can be written

$$(H_0 - H_n)^{1/2} = \frac{\pi}{L}\left(\frac{D}{g\mu_B}\right)^{1/2} n , \tag{18}$$

where $D = 2Sa^2(J_1 + J_2)$ and L is the film thickness. In obtaining Eq. (18), it has been assumed that the magnetization is the same at both fields. For EuS at 1.1 K and $H \sim 23$ kOe ($H_i = 8.6$ kG) this condition is satisfied and results on films with thickness of 258 and 624 Å plotted using Eq. (18) obey this relationship to within the experimental uncertainty. The observation of only odd-n values is expected for complete spin pinning at both surfaces. The existence of a small intercept obtained from a least-squares analysis of the data indicated that spin pinning was not complete and that its effect should be considered.

We have extended our measurements to higher temperatures and show in Fig. 3 representative results near the limits of this temperature interval. The logarithm of the absorption is displayed versus field permitting the variation for a large number of spin-wave peaks to be compared on the same plot and was obtained using conventional operational-amplifier techniques. The abrupt termination of the data near 23.6 kOe represents the field limit of our magnet. At 4.2 K, the decrease in the magnetization causes a shift of the FMR. In this figure, we have displaced the magnetic field to superimpose the two FMR peaks illustrating how the SSWR peaks are shifted with respect to each other as the temperature is changed. The 1-kOe field interval over which peaks are observed is sufficiently large such that the assumption of a field-independent magnetization used in deriving Eq. (18) must be discarded at all but the lowest temperatures. The field dependence of the demagnetizing field, $4\pi M$, must

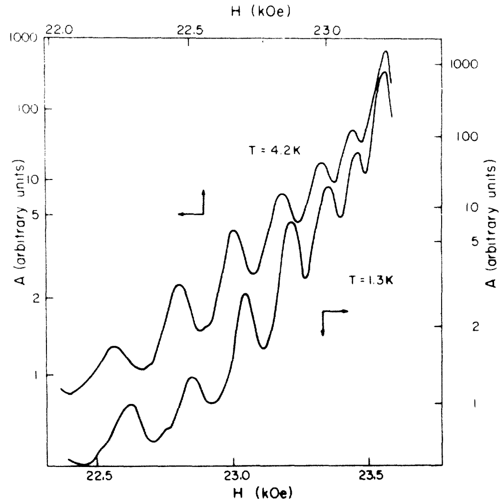

FIG. 3. Logarithm of the SSW absorption versus applied magnetic field at the temperatures $T = 1.3$ and 4.2 K in an EuS film of thickness 624 Å. The magnetic field scales have been displaced so that the first peaks coincide and the change with temperature is more evident.

be calculated and applied as a correction to the observed peak positions for the determination of $D(T)$. This correction is applied and the results compared with theory in Sec. IV C.

We have extended our original analysis, introducing a phenomenological surface-anisotropy energy to represent the boundary conditions. The expression used for the surface-anisotropy energy and the resulting transcendental equation which must be solved are those obtained by Soohoo[25] and Pincus[26] and are given in the Appendix. Analyzing the results obtained on a 624-Å film at 1.2 K gives the results for $(J_1 + J_2)/k_B$ of 84 ± 1 mK when complete pinning is assumed but becomes 77 ± 1 mK when the wave-vector-dependent boundary conditions are taken into account. This uncertainty reflects the accuracy with which the field of individual peaks could be determined and fit to the theoretical curves. The precision of ±1 mK has been quoted to demonstrate the importance of including the surface boundary conditions. The absolute uncertainty is limited by our ability to measure the film thickness and corresponds to ±10 mK.

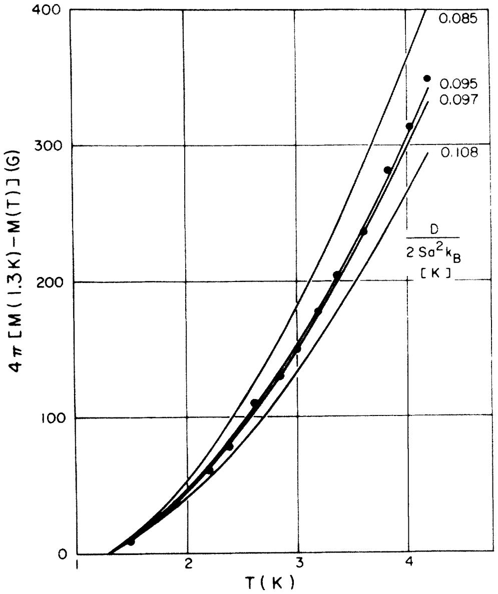

FIG. 4. Experimentally determined change in the magnetization with respect to its value at 1.3 K is shown by open circles plotted versus temperature. The solid curves represent the theoretical change calculated using spin-wave theory, Eq. (10a) for different values of the exchange constant $(D/2Sa^2) = J_1 + J_2$.

B. Magnetization versus temperature

The ferromagnetic-resonance field-frequency relation with H normal to the surface of a thin film, given by

$$\hbar\omega = g\mu_B(H - 4\pi M)\,, \tag{19}$$

can be obtained by setting $k = 0$ in Eq. (3) and is frequently referred to as "the k-equal-zero mode" or as "the uniform precession." This expression, obtained from spin-wave theory, is identical to the classical result first obtained by Kittel.[27] The spin-wave dispersion relation, Eq. (3), is not exact in the limit of very small k due to the long range of the dipolar interaction. For $k \approx 0$ the magnetostatic modes become important and couple to the spin wave or "exchange modes."[28] The resulting mode interaction leads to a substantial modification in the dispersion relations which becomes important when the transverse and longitudinal film dimensions are comparable. These corrections and that due to the bulk anisotropy energy in the interval 1-4 K[23] represent a negligible correction to Eq. (19) and can be ignored.

The magnetization as a function of temperature has been determined from the temperature dependence of the FMR at constant frequency. The results obtained are shown in Fig. 4 plotted as the magnitude of the decrease in magnetization from its value at 1.3 K, $[M(1.3) - M(T)]$, versus temperature. Plotted in the same figure are theoretical curves of $\mathfrak{M}(H, T)$, Eq. (10a) for various values of the exchange stiffness but with $H_i = 8.44$ kG ($\nu = 23.63$ GHz). The best agreement is very sensitive to D and is bounded by the values 0.095 and 0.097. The approximation used in the calculation of $\mathfrak{M}(H, T)$ will lead to an additional uncertainty that is difficult to estimate. Our result from the temperature dependence of the magnetization is

$$(J_1 + J_2)/k_B = 96 \pm 3 \text{ mK}.$$

The uncertainty in this quantity represents an estimate of the theoretical contribution due to the approximations made in calculating $\mathfrak{M}(H, T)$. We observe that this determination of $J_1 + J_2$ does not depend on knowing the film thickness.

C. Temperature dependence of the exchange stiffness and surface-anisotropy constants

Using Eq. (5) with $\theta_k = 0$, we obtain the relation between the wave vectors of two SSW peaks and the *internal* fields at which they occur:

$$D(k_n^2 - k_m^2) = g\mu_B[H_i(m) - H_i(n)]\ . \tag{20}$$

At finite temperature the magnetization will change when the applied field is varied giving the following relation between the internal and applied fields:

$$H_i(m) - H_i(n) = [H(m) - H(n)] \times \left\{1 - 4\pi \frac{M[H(m)] - M[H(n)]}{H(m) - H(n)}\right\}\ . \tag{21}$$

We set $m = 1$ in our analysis and determine the internal field differences at each temperature from the measured values of the applied field and by calculating $M(H)$ numerically from spin-wave theory [Sec. II, Eq. (10a)]. The influence of this correction is seen more clearly by introducing the differential susceptibility $\chi = \partial M/\partial H$. For small field differences, Eq. (21) is approximately given by

$$H_i(m) - H_i(n) \cong [H(m) - H(n)][1 - 4\pi\chi(H, T)] .$$

For the internal fields and temperatures over which SSWR results were analyzed, $H = 7.4-8.4$ kOe, $T/T_C \leq 0.25$, a constant value of the demagnetizing field was used in calculating M from spin-wave theory giving a negligible error for the internal field differences. Similarly, the predicted field dependence of D at each temperature, Eq. (17b) is negligible over the range of internal fields encountered.

The values of k for which standing spin waves can exist depends on the film thickness L and the ratio of surface-anisotropy to exchange energy, k_c as shown in the Appendix, Eqs. (A2a) and (A2b). We have calculated numerically for odd modes, the spectrum of k values for different values of k_c. For the correct value of k_c, a plot of $k_n^2 - k_1^2$ versus the internal field differences [Eq. (20)] is a straight line with zero intercept and a slope equal to $1/D(T)$. This procedure leads to an unambiguous value of k_c which corroborates our indexing of the observed peaks. For a different peak-index assignments, no values of k_c could be found which fit the experimental results.

Our results presented as $D(T)/D_0$ and H_s as a function of temperature are shown in Fig. 5. The exchange stiffness, $D(T)$, changes less than 1% while the surface-anisotropy field decreases rapidly with increasing temperature. From our analysis we find that the observed change in the separation between spin-wave peaks with temperature, Fig. 3, arises principally from the field and temperature dependence of the internal field and temperature dependence of the surface-anisotropy energy. The surface anisotropy field of −1.7 kG at 1.3 K is to be compared with the bulk values at the same temperature, $K_1/M = -30$ G and $K_2/M = 12$ G.[23] In this temperature interval, H_s decreases by a factor of 5 while the bulk K_1/M and K_2/M decrease by 10% and 40%, respectively. This negative surface-anisotropy energy is of the easy plane type. We have found that with the increased separation between peaks in the thinnest films, even modes could be resolved but that their intensity was much weaker than the odd series. An example of such an even peak is shown at ~22 kOe in Fig. 6 for one of our thinnest films. At this temperature, spin pinning should be very weak and any differences between the two film surfaces should be readily apparent. The weakness of this even mode leads us to conclude that the two surfaces are essentially equivalent. The large value of the anisotropy field and its rapid decrease with temperature is clearly a unique surface property and is relatively insensitive to the materials with which the EuS is in contact.

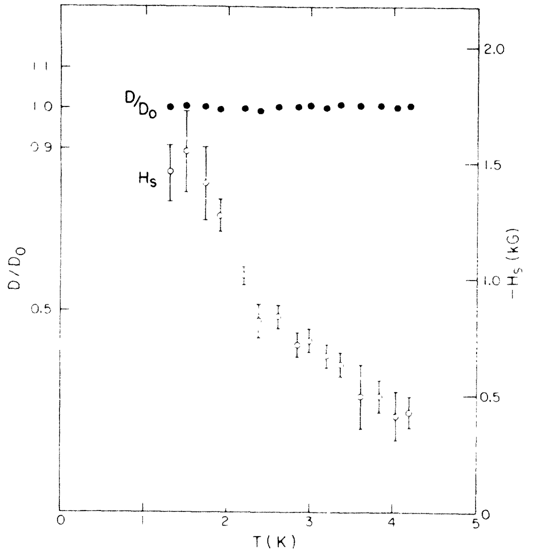

FIG. 5. Experimentally determined variations of the exchange constant plotted as $D(T)/D_0$ and the surface-anisotropy field H_s versus temperature.

In Sec. II, we calculated $D(T)$ for two crystallographic directions. Since thick-film x-ray dif-

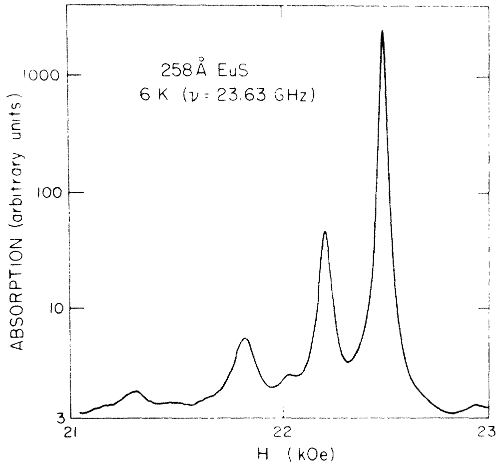

FIG. 6. SSW absorption spectrum of a 258-Å film measured at 6 K. The strongest peak corresponds to ferromagnetic resonance with the major peaks at lower fields at the SSW modes $n = 1, 3$, and 5, respectively. The weak peak between 1 and 3 at 22 kOe corresponds to $n = 2$.

fraction shows a dominance of [111] plane formation during deposition, we will regard the films as growing preferentially with a ⟨111⟩ direction normal to the film surface and parallel to the applied magnetic field. The expression for the change $\Delta D(T)$, Eq. (17b), can be written

$$\frac{\Delta D(T)}{D_0}=-(J_1+2J_2)D(J_1+J_2,H,T)\ . \tag{22}$$

A comparison with theory has been made using the value of $J_1+J_2=96$ mK obtained from the magnetization results, Sec. IV B, to calculate $\Delta D(T)/D_0$. A plot of the experimental results versus theory gives a straight line, the slope of which determines the coefficient $-(J_1+2J_2)$. A least-squares analysis of the data is shown in Fig. 7 by the solid line and corresponds to the ratio $J_2/J_1=-0.59\pm0.03$. We have also analyzed the data using values of J_1+J_2 determined from the standing-spin-wave resonance, Sec. IV A, obtaining $J_2/J_1=-0.55\pm0.03$. In the latter case we have averaged the results of several different films prepared at different times but with identical procedures. The uncertainty given is that obtained by fitting with the average value of J_1+J_2 and does not represent the absolute uncertainty of this quantity. Because the ratio J_2/J_1 is relatively insensitive to the exact value of J_1+J_2 for $J_2/J_1\sim-\frac{1}{2}$ and with the large scatter in $\Delta D(T)$, we take the average of these two determinations obtaining

$$J_2/J_1=-0.57\pm0.05\ .$$

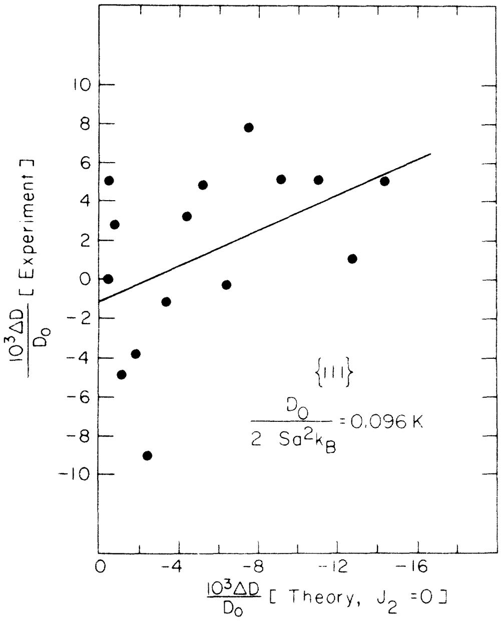

FIG. 7. Plot of the experimental versus theoretical values of $\Delta D(T)/D_0$ calculated for H_i parallel to a ⟨111⟩ direction, Eq. (17b). The solid line represents the least-squares determination of the linear relation between theory and experiment. The scale has been expanded to show the near temperature independence of $\Delta D/D_0$ when $J_2/J_1\sim-\frac{1}{2}$. The weakness of the temperature dependence of $\Delta D/D_0$ and scatter do not permit a distinction between the assumed ⟨111⟩ orientation of the film and the predictions for a ⟨100⟩ film, see Figs. 1 and 2 for $J_2/J_1\sim-\frac{1}{2}$.

Our result for J_2/J_1, treating the films as being single rather than polycrystalline, is based on our x-ray diffraction results. Even though the scatter in $\Delta D(T)$ is large, our results indicate that $D(T)$ *increases* with increasing temperature. Our calculation for a (100) plane, Fig. 1, suggests that the observed increase would also result for a polycrystalline film only if $|J_2|/J_1\geq0.5$. Similarly, the spin-wave dispersion relation including k^4 terms is

$$\begin{aligned}\hbar\omega_k=&\,g\mu_BH_i+2S(J_1+J_2)a^2k^2\\&-2S[\tfrac{2}{5}(J_1+2J_2)](\tfrac{1}{2}a)^4k^4\\&-2S[\tfrac{1}{12}(J_1-8J_2)\Gamma(\theta,\varphi)](\tfrac{1}{2}a)^4k^4\end{aligned} \tag{23}$$

where

$$\Gamma(\theta,\varphi)=\sin^4\theta\sin^2 2\varphi+\sin^2 2\theta-\tfrac{4}{5}\ .$$

The lack of experimental evidence of a k^4 contribution to the dispersion relation is consistent with either the assumption of polycrystalline films *and* $J_1+2J_2\cong0$ or single-crystal films with their surface normal along a ⟨111⟩ direction. Lastly, we found, Sec. IV B, that the decrease in the magnetization with increasing temperature is described very well by a spin-wave calculation up to T/T_C ~0.25 with magnon-magnon interactions neglected. In zero magnetic field, these interactions contribute a term in $T^{5/2}$ with the coefficient proportional to J_1+2J_2.[18] The *apparent* absence of a $T^{5/2}$ contribution to $M(H,T)$ is consistent with our value of J_2/J_1 when magnon-magnon interactions *are* included.

We use the results obtained for J_1+J_2 and J_2/J_1 to determine J_1 and J_2 separately. Since the magnetization and the SSWR results give independent determinations of J_1+J_2, we have weighted the two results in inverse proportion to the estimated absolute uncertainty and obtain the following set of parameters for EuS:

$$J_1+J_2=92\pm3\ \text{mK},\quad J_2/J_1=-0.57\pm0.05\ ;$$

and

$$J_1=214\pm26\ \text{mK},\quad J_2=-122\pm25\ \text{mK}\ .$$

V. DISCUSSION AND CONCLUSIONS

The results of this work are presented in Table I. The values of J_1, J_2, and θ_p are calculated for ease of comparison with other experiments. In presenting uncertainties in the quantities $J_1 + J_2$ and J_2/J_1 calculated from the work of others, we have assumed those quoted for J_1 and J_2 to be independent, and recognize that this probably is an over estimate. With the exception of the Green's-function interpretation, the various experimental results for J_1 are in agreement. The values of J_2 show a wider variation and are not consistent within the quoted uncertainties. Our results and those of inelastic neutron scattering are in accord for J_2, however, the ratio J_2/J_1 represents a lack of agreement between the two experiments which we feel is significant.

The uncertainties in the values of J_1 and J_2 obtained by Charap and Boyd[12] are, we suspect, too low. They point out specifically that their analysis, using J_1, J_2, and H_a as adjustable parameters, did not give a "unique" best set. They state that a reasonable fit to their data could be obtained for the ratio $-J_2/J_1 = p$ in the interval $-0.3 < p < 0.6$. Their reanalysis of the specific-heat data of McCollum and Callaway[11] reduced this interval to $0.2 < p < 0.6$ The bulk magnetization for their sample was 5–8% lower than expected probably indicating magnetic dilution due to Eu^{3+} ions being present to this extent. They took this "dilution" into account, reducing $4\pi M_0$ by this percentage in their analysis. We suggest, tentatively, that their observed anisotropy field could be understood as a surface anisotropy at an interface between EuS and the "nonmagnetic dilutant" presumed (by us) to be Eu_2O_3. This interpretation, assuming a definite segregation between magnetic and nonmagnetic regions, suggests that the theoretical value of $4\pi M_0$ would be more appropriate in their analysis. The results of Passenheim *et al.*[14] do not include an estimate of uncertainties in J_1 and J_2, but as noted in Sec. I, their values give a progressively worse fit to the data with increasing applied fields. We conclude that these additional uncertainties in the interpretation of magnetization and specific-heat data would account for the apparent disagreement with values of J_1 and J_2 obtained in this work.

There appears to be a real disagreement between our results and those of Passell *et al.*[16] Both SSWR and neutron scattering experiments are directly related to the spin-wave dispersion relations but being most sensitive to regions near the center and surface of the BZ, respectively. The discrepancy between the two experiments could be an indication that (i) magnon-magnon interactions (neglected in Ref. 16) make a significant contribution to spin-wave energies near the BZ boundaries at finite temperatures, and (ii) that the treatment of the dipolar interaction in the continuum approximation, as pointed out by Keffer,[18] is inadequate for the large wave vectors encountered in neutron scattering.

The last entries in Table I have been obtained by Swendsen[29] using a Green's-function theory and his

TABLE I. Results of the present work.

	$(J_1 + J_2)/k_B$ (K)	J_2/J_1	J_1/k_B (K)	J_2/k_B (K)	Θ_p (K)[a]
SSWR absorption[b]	(0.092 ± 0.003)	$-(0.57 \pm 0.05)$	(0.214 ± 0.026)	$-(0.122 \pm 0.025)$	(19.3 ± 3.6)
Inelastic neutron scattering[c]	(0.136 ± 0.021)[d]	$-(0.42 \pm 0.07)$[d]	(0.234 ± 0.016)	$-(0.098 \pm 0.014)$	(23.3 ± 2.2)
High-temperature EPR linewidth[e]	(0.08 ± 0.04)[d]	$-(0.56 \pm 0.23)$[d]	(0.18 ± 0.02)	$-(0.10 \pm 0.04)$	(16.4 ± 3.6)
Magnetization vs temperature, $H = 0$[f]	(0.12 ± 0.02)[d]	$-(0.4 \pm 0.1)$[d]	(0.20 ± 0.01)	$-(0.08 \pm 0.02)$	(20.2 ± 1.8)
Specific heat (with and without applied field)[g]	0.14[d]	−0.3[d]	0.20	−(0.06)	21.4
Green's function theoretical analysis using T_c and θ_p [h]	0.134[d]	−0.113	0.151	−0.017	19

[a] Calculated using $\Theta_p = 126J_1 + 63J_2$.

[b] This work.

[c] Reference 16.

[d] Calculated by us from the quoted results. The uncertainties have also been calculated and may be too high.

[e] A. A. Samokhvalov and V. S. Babushkin, Fiz. Tverd. Tela 12, 13 (1970) [Sov. Phys.-Solid State 12, 9 (1970)].

[f] Reference 12.

[g] Reference 14.

[h] Reference 29.

interpretation of the ferromagnetic and paramagnetic Curie temperatures. His results for J_1 and J_2, but particularly the ratio J_2/J_1, do not agree with the other experimental results. Swendsen treats magnon-magnon interactions and shows that the renormalization for each "exchange shell," i.e., J_1 and J_2 contributions, is different. Our calculation of magnon-magnon-interaction contribution using spin-wave theory gives a similar result, Eqs. (17a) and (17b). The role of J_2/J_1 in determining the temperature dependence of the effective exchange constant $D(T)$ is shown explicitly in Figs. 1 and 2. In the low-temperature limit and near the center of the BZ, it is expected that spin-wave and Green's-function results will be in agreement. Also the two theories will reduce to the exact "single-magnon" excitation spectrum in the $T=0$ K limit. Our results are most sensitive to the spin-wave dispersion relations near the center of the BZ where the approximate treatment of the dipolar and magnon-magnon interactions should be most reliable. We conclude that the disagreement with Swendsen's analysis represents a question as to the correctness of his equation used to calculate the ferromagnetic Curie temperature.

ACKNOWLEDGMENTS

We would like to thank Professor A. W. Lawson for his encouragement and many helpful conversations during the course of this work. The assistance of G. S. Cooper, B. Cutler, A. Kasuya, and R. Ketchum is gratefully acknowledged. Our particular thanks are extended to T. M. Kwon for his assistance.

APPENDIX: SURFACE BOUNDARY CONDITIONS

Soohoo[25] and Pincus[26] have considered the effect of surface boundary conditions and their effect on the values of the wave vector k in a SSWR experiment. While the introduction of an "effective-surface-anisotropy field" by Pincus leads to the same result as obtained by Soohoo, we prefer the description in terms of a "surface-anisotropy energy."

Soohoo treats the surfaces of a magnetic thin film by the introduction of a phenomenological uniaxial surface-anisotropy energy given by

$$E_s(\theta, \varphi) = K_s \cos^2\theta \;, \tag{A1}$$

where θ is the angle between the magnetization and the symmetry axis which we choose as being normal to the film surface. For convenience, the results are expressed in terms of K_s' given by $K_s \equiv K_s' a$, where a is the lattice parameter. In the general case, K_s would be different at the two surfaces. For the magnetic field normal to the film and identical surface-anisotropy energies, the standing-wave solutions are determined by solving the equation:

$$\tan kL = \frac{-2}{k/k_c - k_c/k} \tag{A2a}$$

where

$$k_c a = \frac{g\mu_B H_s}{2S(J_1+J_2)} \;, \tag{A2b}$$

$$H_s = aK_s'/M \,. \tag{A2c}$$

In these expressions, L is the film thickness and the other terms have their usual meaning. Positive values of K_s' correspond to easy-plane anisotropy with a "hard axis" normal to the film surface and negative values to an easy axis in the film normal direction. The equation for k has two sets of solutions which can be written

$$\tan\frac{kL}{2} = \frac{k}{k_c}\ \text{(odd)}, \quad \cot\frac{kL}{2} = -\frac{k}{k_c}\ \text{(even)}\;, \tag{A3}$$

where the notation even and odd refers to whether the solutions correspond to antinodes or nodes at the midpoint between the two surfaces, respectively. For insulating films in a spatially uniform rf exciting field (the conditions of this experiment) only the even modes will be excited. In the limit of "complete pinning," $k/k_c \ll 1$, the observed SSW peaks are given by

$$k_n = (2n+1)\pi/L \;,$$

where $n = 1, 2, \ldots$. In our initial observation of SSWR at 1.1 K, we found the results to be well described by "complete pinning."[15] In the present work, the values of k were determined by varying k_c in the expression for even modes to obtain a fit to our data at each temperature. The change of H_s with temperature (Fig. 5) causes a variation from "nearly complete" to "weak" spin pinning with increasing temperature. Also, the "pinning" is a function of wave vector and is different for different modes with a finite nonzero value of the surface-anisotropy energy.

The use of a simple surface-anisotropy energy to describe the surface spin pinning has encountered difficulties when applied to metals. Kooi *et al.*[30] reviewed the experimental situation in 1964 and described various alternate descriptions of spin pinning that more nearly described experimental results. They concluded, "... that it is very difficult to determine exchange parameters by means of spin wave resonance in films." Subsequent experimental work[31] has shown that when metallic films are prepared with special attention to substrate cleanliness and very-high-vacuum conditions employed, the problems of interpretation described by Kooi *et al.* diminish due to the reduction of antiferromagnetic oxide formation during evaporation. These problems do not arise in the preparation of EuS films. The saturation magnetization of our films and bulk properties (ferromagnetic-resonance linewidth) of material prepared by vacuum fractional distillation indicate that oxide formation (EuO or Eu_2O_3) and/or molecular dissociation do not occur to an observable extent. A considerable amount of theoretical work has been directed to the problem of surface effects and, in particular, the coupling between "exchange" and "magnetostatic" spin-wave modes.[28,32] The results indicate that for very thin films having large transverse dimensions and in the field-normal geometry, this coupling makes a negligible contribution to the "exchange only" description. We conclude this discussion by noting that EuS represents a system for which a simple surface-anisotropy-energy model is applicable. The relative insensitivity of this anisotropy energy to the composition of nonmagnetic substrates would provide a unique opportunity to study surface anisotropy through measurements of the influence of the interface between two magnetic materials on the "surface" pinning.

---

*Work supported primarily by the U. S. Atomic Energy Commission and is Report No. UCR 34 P77-36.

†Present address: Centre Electronique Horloger, CH-2001 Neuchatel, Switzerland.

‡Present address: Dept. of Physics, University of Wisconsin, Milwaukee, Wisc. 53201

§Permanent address: Research Institute for Iron, Steel and Other Metals, Tohoku University, Katahira, Sendai, Japan.

[1] B. T. Matthias, R. M. Bozorth, and J. H. Van Vleck, Phys. Rev. Lett. 7, 160 (1961).

[2] (a) T. R. McGuire, B. E. Argyle, M. W. Shafer, and J. S. Smart, Appl. Phys. Lett. 1, 17 (1962); (b) T. R. McGuire and M. W. Shafer, J. Appl. Phys. 35, 984 (1964).

[3] (a) G. Busch, P. Junod, M. Risi, and O. Vogt, in *Proceedings of the International Conference on Semiconductors, Exeter* (The Institute of Physics and The Physical Society, London, 1962), p. 727. (b) G. Busch, J. Appl. Phys. 38, 1386 (1964).

[4] S. van Houten, Phys. Lett. 2, 215 (1962); U. Enz, J. F. Fast, S. van Houten, and J. Smit, Philips Res. Reports 17, 451 (1962).

[5] R. L. Wild and R. D. Archer, Bull. Am. Phys. Soc. 7, 440 (1962).

[6] S. Methfessel and D. C. Mattis, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1968), Vol. XVIII/1, p. 389. The magnetic properties of the insulating europium chalcogenides together with the possible nature and origin of the observed exchange parameters are discussed on pp. 505–512. The applicability of a nearly "ideal" Heinsenberg model and experimental results interpreted on the basis of molecular-field and spin-wave theories are summarized.

[7] (a) N. Miyata and B. E. Argyle, Phys. Rev. 157, 448 (1967). (b) Richard S. Hughes, Ph.D. thesis (University of California, Riverside, 1970, unpublished).

[8] (a) S. von Molnar and A. W. Lawson, Phys. Rev. 139, A1598 (1965). (b) Glen E. Everett and R. A. Ketcham, J. Phys. 32, C1-545 (1971).

[9] D. E. Eastman, Phys. Lett. A 28, 136 (1968).

[10] Atsuo Kasuya and M. Tachiki, in *Proceedings of the Seventeenth Annual Conference on Magnetism and Magnetic Materials, Chicago, 1971*, edited by C. D. Graham, Jr. and J. J. Rhyne (AIP, New York, 1971), p. 845.

[11] Joseph Callaway and D. C. McCollum, Phys. Rev. 130, 1741 (1963); D. C. McCollum and J. Callaway, Phys. Rev. Lett. 9, 376 (1962).

[12] S. H. Charap and E. L. Boyd, Phys. Rev. 133, A811 (1964).

[13] T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).

[14] B. C. Passenheim, D. C. McCollum, Jr., and J. Callaway, Phys. Lett. 23, 634 (1966).

[15] P. K. Schwob and Glen E. Everett, J. Phys. 32, C1-1066 (1971).

[16] L. Passell, O. W. Dietrich, and J. Als-Nielsen, Ref. 10, p. 1251.

[17] P. W. Anderson and H. Suhl, Phys. Rev. 100, 1788 (1955).

[18] F. Keffer, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1966), Vol. XVIII/2, p. 49, Eq. (17.4). This result is for an ellipsoid with orthogonal principal axes with H parallel to one of them. The range of k for which this equation is useful and why it is not exact in the $k=0$ limit is also discussed.

[19] Reference 18, p. 47; also, Sec. 7, p. 18. The existence of bound pairs of spin-wave states and other corrections necessary for a reliable description of spin-wave energies when k is near a BZ boundary are discussed.

[20] The reduction in the magnetization due to zero-point fluctuations is small in ferromagnets with its effect being largest in zero field. With increasing field these fluctuations are suppressed by the field becoming completely negligible at low temperatures and high fields.

[21] T. Oguchi, Phys. Rev. 117, 117 (1960).

[22] W. Marshall, in *Proceedings of the Eighth International Conference Low Temperature Physics, London, 1962*, edited by R. O. Davies (Butterworths, London, 1963).

[23] G. E. Everett and R. A. Ketcham, J. Phys. 32, C1-545 (1971). The dominant contribution to the magnetocrystalline anisotropy energy is from a single-ion mechanism, hence bulk and thin-film "intrinsic" anisotropy energies would not be expected to be greatly different, particularly for the substrate temperatures used.

[24] Y. Shapira and T. B. Reed, in Ref. 10, p. 837.

[25] R. F. Soohoo, *Magnetic Thin Films* (Harper and Row, New York, 1965), Sec. 11.3, p. 231.

[26] P. Pincus, Phys. Rev. 118, 658 (1960).

[27] C. Kittel, Phys. Rev. 73, 155 (1948).

[28] (a) T. Wolfram and R. E. DeWames, Phys. Rev. B 1, 4358 (1970). (b) M. Sparks, Phys. Rev. B 1, 3831 (1970).

[29] R. H. Swendsen, Phys. Rev. B 5, 116 (1972).

[30] C. F. Kooi, P. E. Wigen, M. R. Shanabarger, and J. V. Kerrigan, J. Appl. Phys. 35, 791 (1964).

[31] Glen C. Baily, J. Appl. Phys. 41, 1012 (1970); G. I. Lykken and D. R. Schmitz, J. Appl. Phys. 41, 1024 (1970); A. Stankoff and A. T. Berteaud, J. Phys. 32, C1-562 (1971).

[32] The following references are given to indicate the activity in this area. The reader is referred to these papers for other references. M. Sparks, Phys. Rev. B 1, 3856 (1970); Phys. Rev. B 1, 3869 (1970); R. E. DeWames and T. Wolfram, J. Appl. Phys. 41, 987 (1970); J. C. Levy, E. Ilisca, and J. L. Motchane, Phys. Rev. B 5, 1099 (1972).